

09/787157

PATENT

Practitioner's Docket No. 55710TRANSMITTAL LETTER TO THE U.S. DESIGNATED OFFICE (DO/US)  
ENTRY INTO THE U.S. NATIONAL STAGE UNDER CHAPTER I

<u>PCT/IP00/04610</u>	<u>July 11, 2000</u>	<u>July 14, 1999</u>
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED

PRESSURE-SENSITIVE ADHESIVE SHEET FOR SURFACE PROTECTION  
TITLE OF INVENTION

Mikihiro ENDO; Syuji ICHIMURA; Kazuhiro KONO; Yoshinaga TSUZUKI  
APPLICANTS

Box PCT  
Assistant Commissioner for Patents  
Washington D.C. 20231  
ATTENTION: DO/US

NOTE: The completion of those filing requirements that can be made at a time later than 20 months from the priority date results from the Commissioner exercising his judgment under the authority granted under 35 U.S.C. § 371(d). The filing receipt will show the actual date of receipt of the last item completing the entry into the national phase. See 37 C.F.R. § 1.491, which states: "An international application enters the national stage when the applicant has filed the documents and fees required by 35 U.S.C. § 371(c) within the periods set forth in § 1.494 and § 1.495."

WARNING: Where the items are those that can be submitted to complete the entry of the international application into the national phase subsequent to 20 months from the priority date, the application is still considered to be in the international stage. And if mailing procedures are utilized to obtain a date the express mail procedure of 37 C.F.R. § 1.10 must be used (because international application papers are not covered by an ordinary certificate of mailing. 37 C.F.R. § 1.8(2)(xi)).

WARNING: Documents and fees must be clearly identified as a submission to enter the national stage under 35 U.S.C. § 371, otherwise the submission will be considered as being made under 35 U.S.C. § 111. 37 C.F.R. § 1.494(f).

---

CERTIFICATION UNDER 37 C.F.R. § 1.10\*  
(Express Mail label number is **mandatory**.)  
(Express Mail certification is optional)

I hereby certify that this paper, along with any document referred to, is being deposited with the United States Postal Service on this date March 14, 2001, in an envelope as "Express Mail Post Office to Addressee," mailing Label Number EL196831915US, addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231.

Laura M. McGuire  
(type or print name of person mailing paper)

  
Signature of person mailing paper

WARNING: Certificate of mailing (first class) or facsimile transmission procedures of 37 C.F.R. § 1.8 cannot be used to obtain a date of mailing or transmission for this correspondence.

\*WARNING: Each paper or fee filed by "Express Mail" must have the number of the "Express Mail" mailing label placed thereon prior to mailing. 37 C.F.R. § 1.10(b).  
"Since the filing of correspondence under § 1.10 without the Express Mail mailing label thereon is an oversight that can be avoided by the exercise of reasonable care, requests for waiver of this requirement will not be granted on petition." Notice of Oct. 24, 1996, 60 Fed. Reg. 56,439, at 56,442.

(Transmittal Letter to the United States Designated Office (DO/US - Entry into National Stage  
under 35 USC 371--page 1 of 7)

**WARNING:** Failure to pay the national fee within 20 months from the priority date will result in the abandonment of the application. The time for payment of the basic fee is not extendable M.P.E.P. § 1893.01(a)(1), 6th ed., rev. 3.

1. Applicant herewith submits to the United States Designated Office (DO/US) the following items under 35 U.S.C. 371:

- a. ☒ This express request to immediately begin national examination procedures (35 U.S.C. § 371(f)).
- b. ☒ The U.S. National Fee (35 U.S.C. § 371(c)(1)) and  
☒ other fees (37 C.F.R. § 1.492), as indicated below:

2 Fees					
CLAIMS FEE	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
*	TOTAL CLAIMS	11 - 20 =	0	x\$ 18.00=	\$0
	INDEPENDENT CLAIMS	3 - 3 =	0	x\$ 78.00=	\$0
	MULTIPLE DEPENDENT CLAIMS(S) (if applicable) + \$260.00				\$ 270.00
BASIC FEE**	The international search fee, as set forth in § 1.445(a)(2) to be paid to the US PTO acting as an international Searching Authority:  <input type="checkbox"/> has been paid (37 CFR 1.492(a)(2)).....\$760.00 <input type="checkbox"/> has not been paid (37 CFR 1.492(a)(3)).....\$970.00  <input checked="" type="checkbox"/> where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 CFR 1.492(a)(5)) ..... \$860.00				\$ 860.00
	Total of above Calculations				= \$1130.00
SMALL ENTITY	Reduction by ½ for filing by small entity, if applicable. Affidavit must be filed also. (note 37 CFR 1.9, 1.27, 1.28)				-
	Subtotal				\$1130.00
	Total National Fee				\$1130.00
	Fee for recording the enclosed assignment document \$40.00 (37 CFR 1.21(h)). (See Item 10 below). See attached "ASSIGNMENT COVER SHEET (37 CFR 3.34)".				\$ 40.00
TOTAL	Total Fees enclosed				\$1170.00

(Transmittal Letter to the United States Designated Office (DO/US - Entry into National Stage under 35 U.S.C. 371--page 2 of 7)

09/787157

JCOB Rec'd PCT/PTO 14 MAR 2001

**\*\*WARNING:** "To avoid abandonment of the application, the applicant shall furnish to the United States Patent and Trademark Office not later than the expiration of 20 months from the priority date; \*\*\* (2) the basic national fee (see § 1.492(a)). The 20-month time limit may not be extended." 37 C.F.R. § 1.494(b).

- i. ☒ A check in the amount of \$ 1170.00 to cover the above fees is enclosed.  
 ii. ☐ Please charge Account No. \_\_\_\_\_ in the amount of \$ \_\_\_\_\_.  
 A duplicate copy of this sheet is enclosed.

**WARNING:** If the translations of the international application and/or oath or declaration have not been submitted by the applicant within twenty (20) months from the priority date, the applicant will be so notified and given a period of time within which to file the translation and/or oath or declaration in order to prevent abandonment. The payment of the surcharge set forth in § 1.492(e) is required as a condition for accepting the oath or declaration later than twenty (20) months after the priority date. The payment of the processing fee set forth in § 1.492(f) is required for acceptance of an English translation later than twenty (20) months after the priority date. Failure to comply with these requirements will result in abandonment of the application. The provisions of § 1.136 will apply. 37 C.F.R. § 1.494(c).

3. A copy of the International application as filed (35 U.S.C. § 371(c)(2)):
- a. ☒ is transmitted herewith.  
 b. ☐ is not required, as the application was filed with the United States Receiving Office.  
 c. ☐ has been transmitted  
 i. ☐ by the International Bureau. Date of mailing of the application Prom form PCT/IB/308): \_\_\_\_\_.  
 ii. ☐ by applicant on \_\_\_\_\_.  
 Date

**NOTE:** Section 1.494(b) was amended to require that the basic national fee and a copy of the international application must be filed with the Office by 20 months from the priority date to avoid abandonment. "The International Bureau nominally provides the copy of the international application to the Office in accordance with PCT Article 20. At the same time, the International Bureau notifies the applicant of the communication to the Office. In accordance with PCT Rule 47.1, that notice shall be accepted by all designated offices as conclusive evidence that the communication has duly taken place. Thus, if the applicant desires to enter the national stage and applicant has received notice from the International Bureau, applicant need only pay the basic national fee by 20 months from the priority date." [This can now be paid subsequently with a surcharge.] Notice of Jan. 7, 1993, 1147 O.G. 29 to 40, at 35.

4. A translation of the International application into the English language (35 U.S.C. § 371(c)(2)):
- a. ☒ is transmitted herewith.  
 b. ☐ is not required as the application was filed in English.  
 c. ☐ was previously transmitted by applicant on \_\_\_\_\_.  
 Date

5. ☐ Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. § 371(c)(3)):

**NOTE:** The Notice of January 7, 1993 indicates that 37 C.F.R. § 1.494(d) was "amended to clarify the existing practice that PCT Article 19 Amendments must be submitted by 20 months from the priority date, which time may not be extended." This Notice further advises: "Of course, the failure to do so does not result in loss of the subject matter of PCT Article 19 amendments. The applicant may submit that subject matter in a preliminary amendment filed under Section 1.121. In many cases, filing an amendment under Section 1.121 is preferable since grammatical or idiomatic errors may be corrected." 1147 O.G. 29-40, at 35. See item 11(c) below. See also 37 C.F.R. § 1.494(d).

(Transmittal Letter to the United States Designated Office (DO/US - Entry into National Stage under 35 U.S.C. 371--page 3 of 7)

JC08 Rec'd PCT/PTO 14 MAR 2001

- a. ☐ are transmitted herewith.
- b. ☐ have been transmitted
- i. ☐ by the International Bureau. Date of mailing of the amendment (from form PCT/IB/308): \_\_\_\_\_.
- ii. ☐ by applicant on \_\_\_\_\_.
- Date
- c. ☒ have not been transmitted, as
- i. ☐ no notification has been received that the International Search Authority has received the Search Copy.
- ii. ☐ the Search Copy was received by the International Searching Authority, but the Search Report has not yet been issued. Date of receipt of Search Copy from form PCT/ISA/202): \_\_\_\_\_.
- iii. ☒ applicant chose not to make amendments under PCT Article 19. Date of mailing of Search Report (from form PCT/ISA/210): \_\_\_\_\_.
- iv. ☐ the time limit for the submission of amendments has not yet expired. The amendments, or a statement that amendments have not been made, will be transmitted before the expiration of the time limit under PCT Rule 46.1.
6. ☒ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. § 371(c)(3)):
- a. ☐ is transmitted herewith.
- b. ☐ is not required as the amendments were made in the English language.
- c. ☒ has not been transmitted for reasons indicated at point 5(c) above.
7. ☒ An oath or declaration of the inventor including power of attorney (35 U.S.C. § 371(c)(4)) complying with 35 U.S.C. § 115
- a. ☐ was previously submitted by applicant on \_\_\_\_\_.
- Date
- b. ☒ is submitted herewith, and such oath or declaration
- i. ☒ is attached to the application.
- ii. ☐ identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or (c) and 5(b); and states that they were reviewed by the inventor, as required by 37 C.F.R. § 1.70.
- iii. ☐ will follow.

II. Other document(s) or information included:

8. ☒ An international Search Report or Declaration under PCT Article 17(2)(a):
- a. ☒ is transmitted herewith.
- b. ☐ has been transmitted by the International Bureau. Date of mailing from form PCT/IB/308): \_\_\_\_\_.
- c. ☐ is not required, as the application was searched by the United States International Searching Authority.
- d. ☐ will be transmitted promptly upon request.
- e. ☐ has been submitted by applicant on \_\_\_\_\_.
- Date
- f. ☐ is not transmitted, as the international search has not yet issued.

09/787157

JC08 Rec'd PCT/PTO 14 MAR 2001

9. ☒ An Information Disclosure Statement under 37 C.F.R. §§ 1.97 and 1.98:
- a. ☐ is transmitted herewith.  
Also transmitted herewith is (are)  
☐ Form PTO-1449 (PTO/SB/08A and 08B)  
☐ Copies of citations listed
- b. ☒ will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. § 371(c).
- c. ☐ was previously submitted by applicant on \_\_\_\_\_.  
Date
10. ☒ An assignment document is transmitted herewith for recording. A separate  
☐ "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or  
☒ FORM PTO—1595 is also attached.  
☐ Please mail the recorded assignment document to:
- i. ☒ the person whose signature and address appears below.
- ii. ☐ the following:
11. ☒ Additional documents
- a. ☒ Copy of request (PCT/RO/101)
- b. ☒ International Publication No. WO 01/05589
- i. ☐ Specification, claims and drawing
- ii. ☒ Front page only
- c. ☐ Preliminary amendment (37 C.F.R. § 1.121)
- d. ☒ Other:  
**Form PCT/IB/301**  
**Form PCT/IB/304**  
**Form PCT/IB/308**
12. ☒ The above checked items are being transmitted
- a. ☐ before the 18th month publication.
- b. ☒ after publication and the article 20 communication, but before 20 months from the priority date.
- c. ☐ after 20 months (revival).

NOTE: *Petition to revive (37 C.F.R. § 1.137(a) or (b)) is necessary if 35 U.S.C. § 371 requirements are submitted after 20 months.*

13. ☐ Certain requirements under 35 U.S.C. § 371 were previously submitted by the applicant on \_\_\_\_\_ namely:  
Date

#### AUTHORIZATION TO CHARGE ADDITIONAL FEES

**WARNING:** *Accurately count claims, especially multiple dependent claims, to avoid unexpected high charges if extra claims are authorized.*

(Transmittal Letter to the United States Designated Office (DO/US - Entry into National Stage under 35 U.S.C. 371--page 5 of 7)

**NOTE:** "A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time. An authorization to charge all required fees, fees under § 1.17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission. Submission of the fee set forth in § 1.17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 C.F.R. § 1.136(a)(3).

**NOTE:** "Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts; amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 C.F.R. § 1.26(a).

☒ The Commissioner is hereby authorized to charge the following additional fees that may be required by this paper and during the entire pendency of this application to Account No. 04-1105.

☒ 37 C.F.R. § 1.492(a)(1), (2), (3), and (4) (filing fees)

**WARNING:** Because failure to pay the national fee within 20 months without extension (37 C.F.R. § 1.494(b)(2)), results in abandonment of the application, it would be best to always check the above box

☒ 37 C.F.R. § 1.492(b), (c), and (d) (presentation of extra claims)

**NOTE:** Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must only be paid or these claims cancelled by amendment, prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 C.F.R. § 1.16(d)), it might be best not to authorize the PTO to charge additional claim fees, except possibly when dealing with amendments after final action.

☒ 37 C.F.R. § 1.17 (application processing fees)

☒ 37 C.F.R. § 1.17(a)(1)-(5)(extension fees pursuant to § 1.136(a).

☐ 37 C.F.R. § 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. § 1.311(b)).

**NOTE:** Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance. 37 C.F.R. § 1.311(b).

**NOTE:** 37 C.F.R. § 1.28(b) requires "Notification of any change in status resulting in loss of entitlement to small entity status must be filed in the application . . . prior to paying or at the time of paying . . . issue fee...." From the wording of 37 C.F.R. § 1.28(b): (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.

☒ 37 C.F.R. § 1.492(e) and (f) (surcharge fees for filing the declaration and/or filing an English translation of an International Application later than 20 months after the priority date.

09/787157

Docket No. 55710

JG08 Rec'd PCT/PTO

14 MAR 2001

  
SIGNATURE OF PRACTITIONER

Reg. No.: 33,860

Peter F. Corless

(type or print name of practitioner)

Tel. No.: (617) 523-3400

Dike, Bronstein, Roberts & Cushman  
Intellectual Property Practice Group  
EDWARDS & ANGELL, LLP

130 Water Street

P.O. Address

Customer No.:

Boston, MA 02109

#146713

## SPECIFICATION

Pressure-sensitive adhesive sheet for surface protection

## 5 TECHNICAL FIELD

The present invention relates to a pressure-sensitive adhesive sheet for surface protection which has excellent weathering resistance to undergo neither chalking nor fracture in the substrate at peeling, even after a prolonged outdoor exposure, which develops less corona odor to enable extended operation of applying it, and which can be manufactured inexpensively with reduced manufacturing process, since no anchor coat treating procedure is required.

15

## BACKGROUND ART

Techniques for protecting a top coating surface formed on the surface of a body of a finish-coated car are disclosed, for example, in Japanese Provisional Patent Publication Nos. Hei 8-143830 and Hei 8-143838 and Japanese Patent Application No. Hei 9-322574. These techniques predominantly use application of pressure-sensitive adhesive sheets for protecting directly to coating surfaces. Such pressure-sensitive adhesive sheets each uses as a substrate a polypropylene type film in view of its strength, flexibility, thermal resistance, etc. However, polypropylene type films undergo chalking when subjected to long outdoor exposure to cause dusting at peeling to litter the surfaces of car bodies with white powder particles, disadvantageously. In addition, polypropylene type films develop strong scorching odor due to corona discharge treatment applied to them for securing adhesion with a release agent, an anchor coating agent, a pressure-sensitive adhesive, etc., which remains considerably after pressure-sensitive adhesive sheets are completed. This gives rise to problems of injuring health



of operators that they are suffered from headache and the like induced by such odor if they carry out application for an extended period.

5       It is an objective of the present invention to provide a pressure sensitive adhesive sheet for surface protection which has excellent weathering resistance to undergo neither chalking nor fracture in the substrate at peeling, even after a prolonged outdoor exposure, which develops  
10   less corona odor to enable extended operation of applying it, and which can be manufactured inexpensively with reduced manufacturing process, since no anchor coat treating procedure is required.

15   DISCLOSURE OF THE INVENTION

      The present invention (1) relates to a pressure-sensitive adhesive sheet for surface protection having a three-layered film in which a layer A, a layer B and a layer C have been laminated in this order and a pressure-sensitive adhesive layer on the layer C; wherein the layer  
20   A contains a polyethylene in an amount of at least 60 % by weight based on the total weight of the layer A; the layer B contains a polypropylene type polymer in an amount of at least 50 % by weight based on the total weight of the layer  
25   B; and the layer C contains a hydrogenated styrene/diene type hydrocarbon copolymer in an amount of at least 10 % by weight based on the total weight of the layer C.

      The present invention (2) relates also to the  
30   pressure-sensitive adhesive sheet for surface protection (1), wherein the layer C contains an ultraviolet stabilizer in an amount of not less than 0.1 part by weight per 100 parts by weight of the hydrogenated styrene/diene type hydrocarbon copolymer contained in the layer C and not more  
35   than 0.5 % by weight based on the total weight of the layer C.

The present invention (3) relates also to the pressure-sensitive adhesive sheet for surface protection (1 or 2), wherein at least one layer in the three-layered film contains titanium dioxide, and the three-layered film has a total titanium dioxide content of 5 to 20 % by weight based on the total weight of the three-layered film.

The present invention (4) relates also to the pressure-sensitive adhesive sheet for surface protection (3), wherein the layers containing titanium dioxide all further contain an ultraviolet stabilizer, and each titanium dioxide-containing layer contains the ultraviolet stabilizer in an amount of not less than 0.5 part by weight per 100 parts by weight of the titanium dioxide contained in that layer and not more than 0.5 % by weight based on the total weight of that layer.

The present invention (5) relates also to the pressure-sensitive adhesive sheet for surface protection (1 to 4), wherein the layer A contains an ultraviolet absorber in an amount of 0.05 to 0.5 % by weight based on the total weight of the layer A.

#### BEST MODE FOR CARRYING OUT THE INVENTION

The layer A according to the present invention contains 60 to 100 % by weight of polyethylene based on the total weight of the layer A. Here, the polyethylene used in the layer A preferably has a density of 0.91 to 0.97, a melt flow rate (hereinafter abbreviated as MFR; ASTM D1238) of 0.02 to 30 g/10 minutes (190°C; load: 2.16 kg), more preferably an MFR of 0.1 to 10 g/10 minutes in view of film forming properties, flexibility of resulting adhesive sheet, etc.

Other components that can be contained in the layer A

include, for example, polymers other than polyethylene and additives. Other polymers include a blend of at least one kind of component selected from the group consisting of polypropylenes, ethylene/propylene copolymers, 5 ethylene/propylene/diene type hydrocarbon copolymers, ethylene/butene-1 copolymers, ethylene/ $\alpha$ -olefin copolymers, ethylene/methacrylic copolymers, metal salts of ethylene/methacrylic copolymers, ethylene/methyl (meth)acrylate copolymers, ethylene/ethyl (meth)acrylate 10 copolymers, ethylene/vinyl acetate copolymers, olefin crystal/diene hydrocarbon/olefin crystal block copolymers (including hydrogenated products thereof), styrene/diene type hydrocarbon/olefin crystal block copolymers (including hydrogenated products thereof), styrene/diene type 15 hydrocarbon/styrene block copolymers (including hydrogenated products thereof) and styrene/diene type hydrocarbon random copolymers (including hydrogenated products thereof). Other polymers are preferably those having an MFR of 0.1 to 30 g/10 minutes (190°C; load: 2.16 20 kg) or those having an MFR of 0.5 to 30 g/10 minutes (230°C; load: 2.16 kg) in view of compatibility with the base polymer, film forming properties and the like.

Further, in view of weathering resistance, the layer A 25 preferably contains an ultraviolet absorber in an amount of 0.05 to 0.5 % by weight based on the total weight of the layer A. The ultraviolet absorber includes, for example, benzotriazole compounds which may be used singly or as a blend of two or more kinds of them.

30 The layer B according to the present invention contains a polypropylene type polymer in an amount of 50 to 100 % by weight based on the total weight of the layer B. Here, the polypropylene type polymer to be contained in the 35 layer B includes, for example, homopolypropylenes and ethylene/propylene copolymers (block copolymers or random

copolymers which are generally referred to as block polypropylene or random polypropylenes). Those having an MFR of 0.3 to 10 g/10 minutes (230°C; load: 2.16 kg) are preferred in view of film forming properties, as well as, tensile strength and flexibility of the resulting pressure-sensitive adhesive sheet.

The layer B may contain polymers other than polypropylene polymers and additives. Other polymers include a blend of at least one kind of component selected from the group consisting, for example, of polyethylenes, ethylene/propylene/diene type hydrocarbon copolymers, ethylene/butene-1-copolymers, ethylene/ $\alpha$ -olefin copolymers, ethylene/methacrylic copolymers, metal salts of ethylene/methacrylic copolymers, ethylene/methyl (meth)acrylate copolymers, ethylene/ethyl (meth)acrylate copolymers, ethylene/vinyl acetate copolymers, olefin crystal/diene hydrocarbon/olefin crystal block copolymers (including hydrogenated products thereof), styrene/diene type hydrocarbon/olefin crystal block copolymers (including hydrogenated products thereof), styrene/diene type hydrocarbon/styrene block copolymers (including hydrogenated products thereof) and styrene/diene type hydrocarbon random copolymers (including hydrogenated products thereof). Further, those having an MFR of 0.02 to 10 g/10 minutes measured at 190°C under a load of 2.16 kg or those having an MFR of 0.3 to 10 g/10 minutes (230°C; load: 2.16 kg) are preferred in view of compatibility with the base polymer, film forming properties and the like.

30

The layer C according to the present invention contains a hydrogenated styrene/diene type hydrocarbon copolymer in an amount of 10 to 100 % by weight based on the total weight of the layer C. The hydrogenated styrene/diene type hydrocarbon copolymer contained in the layer C is preferably a block copolymer or a random

35

copolymer having an MFR of 0.3 to 10 g/10 minutes (230°C; load: 2.16 kg) and a styrene content of 5 to 40 wt % in view of adhesion with the pressure-sensitive adhesive layer, film forming properties and the like. The layer C  
5 may contain polymers other than the hydrogenated styrene/diene type hydrocarbon copolymer and additives. Other polymers include a blend of at least one kind of component selected from the group consisting, for example, of polyolefin polymers such as polyethylenes,  
10 polypropylenes, ethylene/propylene copolymers, ethylene/propylene/diene type hydrocarbon copolymers, ethylene/butene-1-copolymers, ethylene/ $\alpha$ -olefin copolymers, ethylene/methacrylic copolymers, metal salts of ethylene/methacrylic copolymers, ethylene/methyl  
15 (meth)acrylate copolymers, ethylene/ethyl (meth)acrylate copolymers, ethylene/vinyl acetate copolymers, olefin crystal/diene hydrocarbon/olefin crystal block copolymers (including hydrogenated products thereof) and styrene/diene type hydrocarbon/olefin crystal block copolymers (including  
20 hydrogenated products thereof). Further, those having an MFR of 0.1 to 30 g/10 minutes measured at 190°C under a load of 2.16 kg or those having an MFR of 0.5 to 30 g/10 minutes (230°C; load: 2.16 kg) are preferred in view of compatibility with the hydrogenated styrene/diene type  
25 hydrocarbon copolymer, film forming properties and the like.

The layer C preferably contains an ultraviolet stabilizer in an amount of not less than 0.1 part by weight  
30 per 100 parts by weight of the hydrogenated styrene/diene type hydrocarbon copolymer contained in the layer C and not more than 0.5 % by weight based on the total weight of the layer C. The ultraviolet stabilizer includes, for example, hindered amine compounds and benzoate compounds, and these  
35 compounds may be used singly or as a blend of two or more kinds of them.

In the three-layered film (layer A/layer B/layer C) according to the present invention, it is preferred that one or two of these three layers or all of them contain titanium dioxide. The three-layered film preferably has a total titanium dioxide content of 5 to 20 wt % based on the total weight of the three-layered film. As the titanium dioxide, a surface-inactivated titanium dioxide treated, for example, with alumina or silica which hardly forms radicals under ultraviolet irradiation is preferred in view of weathering resistance. If a surface-activated titanium dioxide is used, radicals are formed on the surface of the titanium dioxide upon ultraviolet irradiation to accelerate deterioration of the substrate layers. Further, the titanium dioxide preferably has a particle diameter of 0.1 to 0.4  $\mu\text{m}$ .

All of the layers containing titanium dioxide preferably contain additionally an ultraviolet stabilizer. It is also preferred that each layer containing titanium dioxide has an ultraviolet stabilizer content of not less than 0.5 part by weight per 100 parts by weight of the titanium dioxide contained therein and not more than 0.5 % by weight based on the total weight of the layer.

The layers A to C may contain various kinds of additives respectively other than those described above, so long as they do not impair the intended objective of the present invention. The additives include, for example, softening agents such as oils, paraffin waxes, epoxy plasticizers and polyester plasticizers; pigments such as titanium dioxide, carbon black and red oxide; fillers such as talc and calcium carbonate; lubricants such as stearic acid, stearyl amide, calcium stearate, barium stearate and zinc stearate; antioxidants such as phenol compounds, phosphite compounds and thioether compounds; ultraviolet

absorbers or stabilizers such as benzotriazole compounds, hindered amine compounds and benzoate compounds; inorganic hollow particles such as glass balloons and silica balloons; microspheres of polymers such as acrylic microspheres and high molecular weight polyolefin powders; and releasability imparting agents such as silicone compounds and long-chain alkyl compounds.

While the pressure-sensitive adhesive layer according to the present invention is not particularly limited, it is preferably a rubber type pressure-sensitive adhesive, particularly preferably at least one kind of component selected from the group consisting of polyisobutylene, butyl rubber and polybutene.

The pressure-sensitive adhesive layer may, as necessary, contain various kinds of additives so long as they do not impair the intended objective of the present invention. The additives include, for example, softening agents such as oils, paraffin waxes, epoxy plasticizers and polyester plasticizers; pigments such as titanium dioxide, carbon black and red oxide; fillers such as talc and calcium carbonate; lubricants such as stearic acid, stearyl amide, calcium stearate, barium stearate and zinc stearate; antioxidants such as phenol compounds, phosphite compounds and thioether compounds; ultraviolet absorbers or stabilizers such as benzotriazole compounds, hindered amine compounds and benzoate compounds; inorganic hollow particles such as glass balloons and silica balloons; microspheres of polymers such as acrylic microspheres and high molecular weight polyolefin powders; and releasability imparting agents such as silicone compounds and long-chain alkyl compounds.

Referring to the thickness of the substrate layers, the layers A, B and C have thickness values of preferably 2

to 200  $\mu\text{m}$ , 2 to 400  $\mu\text{m}$  and 2 to 200  $\mu\text{m}$ , respectively, more preferably 5 to 50  $\mu\text{m}$ , 10 to 100  $\mu\text{m}$  and 5 to 50  $\mu\text{m}$ , respectively. The pressure-sensitive adhesive layer has a thickness of preferably 2 to 100  $\mu\text{m}$ , more preferably 5 to 30  $\mu\text{m}$ .

The pressure-sensitive adhesive sheet for surface protection according to the present invention is prepared by forming a three-layered film consisting of a layer A, a layer B and a layer C. After the film is subjected to one-side corona treatment on the layer A side, the resulting layer A side is treated with a releasing agent, and a pressure-sensitive adhesive layer is then formed on the layer C side by means of solution coating or hot melt extrusion coating. Alternatively, substrate layers and a pressure-sensitive adhesive layer are coextruded to form a layer A/layer B/layer C/pressure-sensitive adhesive layer, followed successively by one-side corona treatment and releasing agent application treatment on the layer A side in a continuous process.

The present invention will be described below specifically by way of nonlimitative examples.

Components used are as follows:

(1) High-density polyethylene (hereinafter abbreviated as "HPE")

HPE-1: HI-ZEX 6800S (high-density polyethylene, manufactured by Mitsui Chemicals, Inc.)

(2) Low-density polyethylene (hereinafter abbreviated as "LPE")

LPE-1: JF630S (low-density polyethylene, manufactured by JAPAN POLYOLEFINS Co., Ltd.)



(3) Polypropylene (hereinafter abbreviated as "HPP")  
HPP-1: IDEMITSU POLYPRO F-200S (polypropylene, manufactured by Idemitsu Petrochemical Co., Ltd.)

5 (4) Ethylene/propylene copolymer (hereinafter abbreviated as "EPP")

EPP-1: NOBLEN BJS-M (ethylene/propylene block copolymer, manufactured by Mitsui Chemicals, Inc.)

10 (5) Ethylene/butene-1 copolymer (hereinafter abbreviated as "EBM")

EBM-1: JSR EBM2011P (ethylene/butene-1 random copolymer, manufactured by JSR Corporation)

15 (6) Hydrogenated styrene/diene type hydrocarbon copolymer (hereinafter abbreviated as "HSR")

HSR-1: DYNARON 1320P (hydrogenated styrene/butadiene random copolymer, manufactured by JSR Corporation)

20 (7) Titanium dioxide (hereinafter abbreviated as "TIO")

TIO-1: TIPAQUE CR90 (rutile type surface-inactivated titanium dioxide, manufactured by ISHIHARA SANGYO KAISYA, LTD.)

25 (8) Ultraviolet stabilizer (hereinafter abbreviated as "HLS")

HLS-1: TINUBIN 622LD (hindered amine ultraviolet stabilizer, manufactured by Ciba Specialty Chemicals)

30 (9) Ultraviolet absorber (hereinafter abbreviated as "UVA")

UVA-1: TINUVIN 326 (benzotriazole ultraviolet absorber, manufactured by Ciba Specialty Chemicals)

35 (10) Antioxidant (hereinafter abbreviated as "AO")

AO-1: IRGANOX 1010 (hindered phenol antioxidant,

manufactured by Ciba Specialty Chemicals)

(11) Polyisobutylene (hereinafter abbreviated as "PIB")

5 PIB-1: VISTANEX MML80 (polyisobutylene, manufactured by  
Exxon Corp.;  $M_{T,v}$ : 990,000)

(12) Polybutene (hereinafter abbreviated as "PB")

10 PB-1: IDEMITSU POLYBUTENE 300R (polybutene, manufactured  
by Idemitsu Petrochemical Co., Ltd.;  $M_n$ : 1330)

Examples 1 to 3 and Comparative Examples 1 to 6

15 Compositions shown in Table 1 were coextruded through  
a T-die extruder to form 60  $\mu$ m-thick three-layered films  
having a ratio of layer A/layer B/layer C = 10 : 40 : 10,  
respectively. Next, each film was subjected to one-side  
corona treatment on the layer A side so that the film may  
have a surface wettability of 50 dyn/cm immediately after  
the treatment, followed by treatment of the same side with  
a long-chain alkyl releasability imparting agent.

20 Subsequently, a pressure-sensitive adhesive containing PIB-  
1/PB-1/UVA-1/AO-1 = 70/30/0.5/0.5 (in terms of weight  
ratio) in a toluene solvent was applied to the layer C side  
to a thickness of 10  $\mu$ m by means of solution coating.

Table 1

	Example 1	Example 2	Example 3	Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5	Comparative Example 6
Layer A	HPE-1 LPE-1 EPP-1 TIO-1 HLS-1 UVA-1 AO-1	70.00 29.80  34.50 0.20 0.15 0.15	70.00 29.70  0.15 0.15	30.00 69.70  0.15 0.15	99.80   0.10 0.10	30.00 69.70  0.15 0.15	99.88   0.02 0.10	30.00 69.70  0.15 0.15	30.00 69.70  0.15 0.15
Layer B	HPP-1 EPP-1 EBM-1 TIO-1 HLS-1 AO-1	60.00 15.00 24.70 0.20 0.10	75.00 24.80 0.10 0.10	75.00 24.80 0.10 0.10	30.00 60.00 9.80 0.10 0.10	30.00 60.00 9.80 0.10 0.10	30.00 60.00 9.80 0.10 0.10	60.00 30.00 9.88 0.02 0.10	67.00 30.00 2.80 0.10 0.10
Layer C	HPP-1 EPP-1 HSR-1 TIO-1 HLS-1 AO-1	40.00 59.80 0.10 0.10	75.00 24.80 0.10 0.10	40.00 59.80 34.65 0.25 0.10	75.00 24.80 0.10 0.10	97.00 2.80 0.10 0.10	75.00 24.80 0.10 0.10	40.00 59.80 0.10 0.10	75.00 24.80 0.10 0.10
Titanium dioxide content in layers A, B and C	16.5%	5.8%	5.8%	6.5%	6.5%	6.5%	6.5%	6.6%	1.9%

\* Thickness: Layer A:Layer B:Layer C = 10 µm:40 µm:10 µm

The pressure-sensitive adhesive sheets for surface protection obtained in Examples 1 to 3 and Comparative Examples 1 to 6 were evaluated in terms of odor, adhesion  
5 between the substrate and adhesive layer, weathering resistance, as well as, application and peeling operability. Odor of the corona-treated films before application of the pressure-sensitive adhesive was also evaluated.

10 (1) Evaluation of odor

Odor of each film immediately after the corona treatment and that of the film after application of the pressure-sensitive adhesive were evaluated.

15 (2) Evaluation of adhesion between substrate and pressure-sensitive adhesive layer

An adhesive cellophane tape (Nichiban Co., Ltd.) was applied to a sample, and they were subjected to T-peeling  
20 at a rate of 0.3 m/min. to evaluate if the pressure-sensitive adhesive layer of the sample came off or not.

(3) Evaluation of weathering resistance

A sample was applied to a poor adherend acrylic paint  
25 film, followed by irradiation with carbon arc. The thus treated sample was then subjected to peeling at a rate of 40 m/min. to evaluate if fracture occurred or not in the substrate and if dusting due to chalking occurred or not during the peeling.

30 (4) Application and peeling operability

Operability when the sample was applied to a finished car coated with a poor adherend acrylic paint and operability when the sample was peeled after 6 month of  
35 outdoor exposure from the application were evaluated.

(5) Remarks

Paint film: poor adherend acrylic coating film (containing silicone) having a wettability (JIS K6768) of lower than 30 dyn/cm.

- 5 Carbon arc irradiation: 1000 hour irradiation using a sunshine weather meter (rainfall: 200 hrs.)

Test result

Test results are shown in Table 2.

Table 2

	Example 1	Example 2	Example 3	Compara- tive Example 1	Compara- tive Example 2	Compara- tive Example 3	Compara- tive Example 4	Compara- tive Example 5	Compara- tive Example 6
Odor	Corona treated film	○	○	×	○	○	○	×	×
	Pressure-sensitive adhesive sheet	○	○	×	○	○	○	×	×
Weathering resistance	Adhesion between substrate and pressure-sensitive adhesive layer	○	○	○	○	×	○	○	○
	Fracture in substrate at peeling	○	○	○	○	○	×	×	×
Application and peeling operability	Chalking	○	○	×	○	○	○	×	×
		○	○	○	×	×	×	○	○

Odor: ○: little odor X: strong odor

Adhesion between the substrate and the pressure-sensitive adhesive layer:

○: no lifting occurred in the adhesive layer

×

Fracture in the substrate at peeling:

○: no fracture occurred X: fracture occurred

Chalking: ○: no white powder dusting occurred X: white powder dusting occurred

Application and peeling operability:

○: excellent

×

As is clear from Table 2, the pressure-sensitive adhesive sheet for surface protection according to the present invention has excellent weathering resistance and can be peeled easily causing neither dusting of white powder particles due to chalking nor fracture in the substrate even when the sheet is peeled speedily after carbon arc irradiation. The sheet of the present invention also has excellent adhesion between the substrate and the pressure-sensitive adhesive layer, enables extended application operation due to reduced corona odor and can be applied and peeled with no difficulty.

# CLAIMS

1. A pressure-sensitive adhesive sheet for surface protection comprising a three-layered film in which a layer A, a layer B and a layer C have been laminated in this order and a pressure-sensitive adhesive layer on the layer C;

wherein the layer A contains a polyethylene in an amount of at least 60 % by weight based on a total weight of the layer A;

the layer B contains a polypropylene polymer in an amount of at least 50 % by weight based on a total weight of the layer B; and

the layer C contains a hydrogenated styrene/diene hydrocarbon copolymer in an amount of at least 10 % by weight of based on a total weight of the layer C.

2. The pressure-sensitive adhesive sheet for surface protection according to Claim 1, wherein the layer C contains an ultraviolet stabilizer in an amount of not less than 0.1 part by weight per 100 parts by weight of the hydrogenated styrene/diene hydrocarbon copolymer contained in the layer C and not more than 0.5 % by weight based on the total weight of the layer C.

3. The pressure-sensitive adhesive sheet for surface protection according to Claim 1 or 2, wherein at least one layer in the three-layered film contains titanium dioxide, and the three-layered film has a total titanium dioxide content of 5 to 20 % by weight based on the total weight of the three-layered film.

4. The pressure-sensitive adhesive sheet for surface protection according to Claim 3, wherein the layers containing titanium dioxide all further contain an ultraviolet stabilizer, and each titanium dioxide-



containing layer contains the ultraviolet stabilizer in an amount of not less than 0.5 part by weight per 100 parts by weight of the titanium dioxide contained in that layer and not more than 0.5 % by weight based on the total weight of that layer.

5. The pressure-sensitive adhesive sheet for surface protection according to any of Claims 1 to 4, wherein the layer A contains an ultraviolet absorber in an amount of 0.05 to 0.5 % by weight based on the total weight of the layer A.

ABSTRACT

The pressure-sensitive adhesive sheet for surface protection has a three-layered film formed by laminating a layer A, a layer B and a layer C in this order and a pressure-sensitive adhesive layer formed on the layer C; wherein the layer A contains a polyethylene in an amount of at least 60 % by weight based on a total weight of the layer A; the layer B contains a polypropylene type polymer in an amount of at least 50 % by weight of based on a total weight of the layer B; and the layer C contains a hydrogenated styrene/diene type hydrocarbon copolymer in an amount of at least 10 % by weight based on the total weight of the layer C. This sheet has excellent weathering resistance to undergo neither chalking nor fracture in the substrate at peeling, even after a prolonged outdoor exposure, develops less corona odor to enable extended operation of applying it, and can be manufactured inexpensively with reduced manufacturing process, since no anchor coat treating procedure is required.

## DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that: My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed at 201) below or an original, first and joint inventor (if plural names are listed at 201-208 below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

# PRESSURE-SENSITIVE ADHESIVE SHEET FOR SURFACE PROTECTION

which is described and claimed in:

- ☐ the specification attached hereto.
- ☐ the specification in U.S. Application Serial Number \_\_\_\_\_, filed on \_\_\_\_\_.
- ☒ the specification in PCT international application Number PCT/JP00/04610  
filed on 11/07/00; and was amended on \_\_\_\_\_.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a). I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.

[illegible]

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s) or PCT international application(s) designating the United States of America that is/are listed below, and, insofar as the subject matter of each of the claims of this application is not disclosed in that/those prior application(s) in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose material information as defined in 37 CFR §1.56(a) which occurred between the filing date of the prior application(s) and the national or PCT international filing date of this application:

Prior U.S. Applications or PCT International Applications Designating the U.S-Benefit Under 35 U.S.C. §120					
U.S. Applications		Status (Check One)			
Application Serial No.	U.S. Filing Date	Patented	Pending	Abandoned	
PCT Applications Designating the U.S.					
Application No.	Filing Date	U.S. Serial No. Assigned			

**CLAIM FOR BENEFIT OF PRIOR U.S. PROVISIONAL APPLICATION(S)**  
(35 U.S.C. § 119(e))

I hereby claim the benefit under Title 35, United States Code, §119(e) of any United States provisional application(s) listed below:

Applicant	Provisional Application Number	Filing Date

**POWER OF ATTORNEY:** As a named inventor, I hereby appoint the following attorney(s) with full powers of association, substitution and revocation to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

Sewall P. Bronstein (Reg. No. 16,919)	Linda M. Buckley (Reg. No. 31,003)	David S. Resnick (Reg. No. 34,235)
David G. Conlin (Reg. No. 27,026)	Ronald I. Eisenstein (Reg. No. 30,628)	Peter F. Corless (Reg. No. 33,860)
George W. Neuner (Reg. No. 26,964)	Henry D. Pahl, Jr. (Reg. No. 20,438)	
Ernest V. Linek (Reg. No. 29,822)	Peter J. Manus (Reg. No. 26,766)	

SEND CORRESPONDENCE TO:	DIRECT TELEPHONE CALLS TO:
Dike, Bronstein, Roberts & Cushman, LLP 130 Water Street Boston, Massachusetts 02109	(617) 523-3400

1-00

2 0 1	FULL NAME OF INVENTOR	LAST NAME <u>ENDO</u>	FIRST NAME <u>Mikihiro</u>	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR FOREIGN COUNTRY <u>Japan JPX</u>	COUNTRY OF CITIZENSHIP <u>Japan</u>
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>C/o NICHIBAN COMPANY LIMITED, 3-3, Sekiguchi 2-chome,</u>	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR COUNTRY AND ZIP CODE <u>112-8663 JAPAN</u>

2-00

2 0 2	FULL NAME OF INVENTOR	LAST NAME <u>ICHIMURA</u>	FIRST NAME <u>Syui</u>	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR FOREIGN COUNTRY <u>Japan JPX</u>	COUNTRY OF CITIZENSHIP <u>Japan</u>
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>C/o NICHIBAN COMPANY LIMITED, 3-3, Sekiguchi 2-chome,</u>	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR COUNTRY AND ZIP CODE <u>112-8663 JAPAN</u>

3-00

2 0 3	FULL NAME OF INVENTOR	LAST NAME <u>KONO</u>	FIRST NAME <u>Kazuhiro</u>	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR FOREIGN COUNTRY <u>Japan JPX</u>	COUNTRY OF CITIZENSHIP <u>Japan</u>
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>C/o NICHIBAN COMPANY LIMITED, 3-3, Sekiguchi 2-chome,</u>	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR COUNTRY AND ZIP CODE <u>112-8663 JAPAN</u>

4-00

2 0 4	FULL NAME OF INVENTOR	LAST NAME <u>TSUZUKI</u>	FIRST NAME <u>Yoshinaga</u>	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR FOREIGN COUNTRY <u>Japan JPX</u>	COUNTRY OF CITIZENSHIP <u>Japan</u>
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>C/o NICHIBAN COMPANY LIMITED, 3-3, Sekiguchi 2-chome,</u>	CITY <u>Bunkyo-ku, Tokyo</u>	STATE OR COUNTRY AND ZIP CODE <u>112-8663 JAPAN</u>

2 0 5	FULL NAME OF INVENTOR	LAST NAME	FIRST NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY AND ZIP CODE

2 0 6	FULL NAME OF INVENTOR	LAST NAME	FIRST NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY AND ZIP CODE

2 0 7	FULL NAME OF INVENTOR	LAST NAME	FIRST NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY AND ZIP CODE

2 0 8	FULL NAME OF INVENTOR	LAST NAME	FIRST NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY AND ZIP CODE

I hereby further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signature of Inventor 201 <i>Mikihiro Endo</i>	Signature of Inventor 202 <i>Syuzi Ichimura</i>
Date: March 6, 2001	Date: March 6, 2001
Signature of Inventor 203 <i>Kazuhiro Kono</i>	Signature of Inventor 204 <i>Yoshimaga Tsuzuki</i>
Date: March 6, 2001	Date: March 6, 2001
Signature of Inventor 205	Signature of Inventor 206
Date:	Date:
Signature of Inventor 207	Signature of Inventor 208
Date:	Date: